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Research Progress on Electrolytes for Intermediate and Low Temperature Solid Oxide Fuel Cells

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Abstract. Reducing the temperature for operating the solid oxide fuel cells (SOFCs) is crucial for advancing their commercialization and large-scale application. With the characteristics of clean and environmentally friendly as well as high energy conversion efficiency, it has been considered as a promising choice for future large-scale power generation devices. However, the drawback of high temperature working condition obstructed its wide utilization. As one of the core components of SOFCs, searching for electrolyte materials which are able to be utilized and modified under relative intermediate and low temperatures is an important method to improving the performance under such temperature conditions. CeO_2-based materials, proton-conducting electrolytes, and semiconductor oxide materials have been widely studied for their high ionic conductivity, low electronic conductivity, and excellent performance after modification. This paper focus on the characteristics and principles of these three types of electrolyte materials, illustrating the advantages and drawbacks of different materials and gives strategies for optimizing material performance in practical applications as well as suggestions for the future development of intermediate and low-temperature electrolytes to further promote the utilization of SOFCs.

Keywords: low temperature, solid oxide fuel cells, electrolytes, performance optimization

1. Introduction

The accelerated global industrialization process has led to the large - scale consumption of fossil fuels. While supporting the rapid development of the social economy, it has further exacerbated the problems of environmental deterioration and energy shortage worldwide. Solid oxide fuel cells (SOFCs) have emerged as a cutting - edge and highly promising technology in the energy field, thanks to their high comprehensive energy conversion efficiency, high stability, long service life, and the environmental advantage of near - zero pollutant emissions.

In practical applications, SOFC technology has demonstrated significant value. SOFC with reduced temperature has a wide range of applications, for example, centralized power plants, household cogeneration, and transportation [1]. The first domestic 25kW SOFC combined heat and power device built and put into operation by Huaqing Jingkun uses industrial by - products as fuel sources, reducing annual carbon dioxide emissions by up to 1200 tons and effectively reducing

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carbon emissions in the industrial sector. However, traditional SOFCs typically operate in the high - temperature range of 800 – 1000 °C. This characteristic poses technical challenges such as excessively long startup times, complex high - temperature sealing and insulation technologies, and high energy consumption of auxiliary systems, which greatly restrict their commercial promotion.

Currently, the bottleneck of SOFCs lies in the high energy required during operation, and the source of this energy is mainly fossil fuels. The combustion of fossil fuels produces CO_2, a greenhouse gas. Excessive CO_2 emissions can lead to a series of environmental problems such as global warming. This is the crux of the problem. At present, humans still largely rely on fossil fuels. Other renewable energy sources either have high costs or produce insufficient energy, and their development is incomplete. Moreover, in a high - temperature environment, the resistance value increases, resulting in significant losses during electricity transmission. As a result, the efficiency of SOFCs is only 50-60%. Additionally, it takes a long time to burn fossil fuels to reach the operating temperature (800 °C) of SOFCs. At such high temperatures, the addition of oxygen may oxidize the metal electrodes, leading to a decrease in efficiency. Furthermore, the cost of metal conductors is high. In recent years, reducing the operating temperature of SOFCs to the medium - low temperature range has become a frontier area of focus for both the academic and industrial communities. Medium - low temperature operation can not only significantly improve the startup performance and operational stability of the battery, optimize the comprehensive energy efficiency of the system, alleviate high - temperature sintering and thermal stress damage of components, and significantly extend the service life of the battery but also reduce the reliance on precious metals and high - temperature - resistant materials and greatly expand its commercial application scenarios.

This paper will systematically review the research progress of medium - low temperature SOFC electrolyte materials and conduct indepth discussions from the aspects of material system innovation, preparation process optimization, performance enhancement mechanisms, and application prospects, aiming to provide theoretical references and technical insights for promoting key breakthroughs and large - scale commercial applications of SOFC technology.

2. Intermediate-temperature SOFC electrolytes

SOFC directly converts the chemical energy in fuels and oxidants into electrical energy through electrochemical reactions. The main products are H_2O and CO_2. If hydrogen is used as fuel, zero carbon emissions can be achieved, making it a truly green energy conversion technology. It has wide fuel adaptability. Not only can it use hydrogen, but it can also be compatible with various fuels such as methane, liquefied petroleum gas, diesel, and industrial by - products. When combined with a waste heat recovery system, it can build an efficient distributed energy supply system.

However, the core bottleneck of traditional SOFC lies in a series of problems caused by high - temperature working conditions. On the one hand, its working temperature needs to reach above 800 °C, and a large amount of fossil fuels are consumed during the heating process. The combustion of fossil fuels produces a large amount of CO_2, and excessive emissions will exacerbate environmental problems such as global warming, which is contrary to the development goal of green energy. On the other hand, in a high - temperature environment, the resistance of the electrolyte increases, resulting in an increase in electrical transmission losses. As a result, the energy conversion efficiency of SOFC is only maintained at 50-60%. In addition, introducing oxygen at high temperatures can easily cause the oxidation of metal electrodes, further reducing the battery efficiency. Moreover, the high cost of metal conductors also limits its large - scale application.

Currently, human beings still rely heavily on fossil fuels. Other renewable energy sources cannot completely replace fossil fuels for the time being due to problems such as high costs, insufficient

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production capacity, or imperfect development, which also exacerbates the application limitations of traditional SOFC. To reduce the working temperature, it is crucial to find a series of electrolyte materials that can work at medium and low temperatures and have high performance. Reducing the working temperature means a decrease in ionic conductivity. To improve the performance of the battery, common methods include reducing the thickness of the electrolyte material to reduce the ohmic internal resistance and finding materials with high ionic conductivity. Electrolyte materials that meet the requirements for medium - and low - temperature operation usually have the following requirements: (1). Good ionic conductivity and low electronic conductivity at medium and low temperatures. (2). Good compactness to prevent the mixing of oxygen and fuel. (3). Good compatibility and interface performance with battery materials. This article mainly lists three types of electrolyte materials that meet the requirements for medium - and low - temperature operation, and discusses their performance and application methods.

2.1. CeO_2 - based electrolytes

The unique properties of CeO_2 stem from its special structure. CeO_2 has a fluorite - type cubic structure, where Ce^{4+} ions are arranged in a face - centered cubic (FCC) structure. Each Ce^{4+} ion is coordinated with eight surrounding O ions, which facilitates the formation of oxygen vacancies, thereby improving the ionic conductivity of the material. Stoichiometric CeO_2 is electronically insulating, so CeO_2-based materials are regarded as excellent choices for medium - and low - temperature solid oxide fuel cell electrolyte materials. However, pure CeO_2 has very low ionic conductivity, with a conductivity of only 1×10^{-5} S/cm at 600°C [2]. Moreover, it cannot form a large number of oxygen vacancies. Therefore, doping with rare - earth metals, transition metals, and non - metallic elements can greatly change the structure and performance of the material to adapt to different application conditions.

However, researchers have found a series of problems with CeO_2 - based materials during testing. During the operation of SOFC, CeO_2 is exposed to fuel gas and a strongly reducing atmosphere, which may reduce Ce^{4+} ions to Ce^{4+} ions. At the same time, the introduction of doping ions may also cause a series of problems, such as an increase in the size of Ce ions, resulting in ion - size mismatch and a decrease in the mechanical properties of the material [3].

2.2. Proton - conducting electrolytes

Among proton - conducting electrolytes, ABO_3 - type perovskite materials (such as $SrZrO_3$ and $BaCeO_3$) are widely used [4]. In their structure, the A - site is usually occupied by +2 valence metal cations, and the B - site is occupied by +4 valence metal cations. The conductivity can be improved by the formation of vacancies due to the loss of oxygen ions in the lattice. These materials need to work at 600 - 1200°C. Another promising material is the $A^{3+}B^{3+}O_3$ - type perovskite, which generates oxygen vacancies by substituting Sr for La, thereby increasing the electrolyte conductivity. Its working temperature is only 600 °C, meeting the requirements for medium - and low - temperature applications.

In addition, research on reducing the working temperature of SOFC by constructing thin - film electrolyte membranes (LT - SOFC structure) has attracted much attention, and this method is expected to be applied on a large scale [5]. The thin - film electrolyte prepared by pulsed laser deposition (PLD) technology can effectively solve the gas cross - over problem caused by the arched structure, reducing the working temperature of SOFC to 450 °C.

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2.3. Semiconductor oxide electrolytes

Using semiconductors as substrates or constructing ionic conductors through doping is a hot research direction for medium - and low - temperature SOFC electrolytes. Semiconductor materials have semiconductor characteristics such as tunable energy band structures. Based on semiconductor materials as electrolytes, researchers have developed SIMFC (Semiconductor - ionic membrane fuel cell) on the basis of SOFC, which uses its semiconductor heterostructure to separate carriers. The specific principle is as follows: The semiconductor on the cathode side and the semiconductor on the anode side will form a p - type semiconductor and an n - type semiconductor under the working atmosphere. Due to the different majority and minority carriers on both sides, a concentration gradient of the two carriers is generated, leading to diffusion. Eventually, a space - charge region is established, and after reaching dynamic equilibrium, a pn junction is formed. The space - charge region (pn junction) will inhibit the movement of electrons, thereby preventing the battery from short - circuiting. At the same time, doping semiconductor materials can also improve the battery performance. For example, an SOFC with a Pt/$SmNiO_3$/Pt structure can achieve an output power of 225 mW cm^{-2} at 500 °C [6]. When Chen et al. prepared the electrolyte using ZnO semiconductor materials, they found that adding 0.5 wt% ZnO could increase the ionic conductivity of YSL by 1.2 times at 800 °C [7]. In addition, H in ZnO is easily ionized to form protons, giving it proton - conduction ability. Moreover, ZnO exhibits excellent thermal stability, oxidation resistance, and optoelectronic properties due to its unique structure [8]. Chen et al. compared the performance of pure ZnO materials and ZnO-LCP composite materials and found that SOFCs based on these two materials achieved power outputs of 482 mW/cm^2 and 864 mW/cm^2 respectively at 550 °C [7]. Among them, the proton conductivity of ZnO - LCP reached 0.05 S/cm at 550 °C, highlighting the great potential of semiconductor materials in electrolyte preparation

3. Optimization strategies

3.1. Element doping

Introducing a small number of heterogeneous elements into materials to modify their electronic and crystal structures is regarded as a common method for improving material performance. The ionic conductivity, sintering performance, and battery power density of materials can be effectively optimized by providing additional carriers or regulating interatomic forces through dopants. Previous studies have illustrated that the radius of doped ions has a great impact on the ionic conductivity of materials. Generally, higher the radius of the doped ions matches it is of host ions, the smaller the lattice strain generated. For example, CeO_2-based electrolytes, after doping with Gd and Sm, the conductivity of the material can reach 0.08 S/cm at 600 °C [9], which is attributed to the appropriate radius matching between doped ions and Ce^{4+}, effectively reducing the hindrance of lattice distortion to ion migration. In addition, the synergistic effect of double doping and multi-doping can significantly enhance the density and ionic conductivity of materials. Li et al. utilized F^- and Ca^{2+} co-doping in GDC (Gd-doped CeO_2), which could both enhanced the sintering performance of the material and the oxygen ion conduction capability [7].

In recent years, the influence of doping ion size on materials has been discussed by many researchers. For example, Mahato et al. the effect of trivalent cation doping on the ionic conductivity of CeO_2, and found that the migration barrier of oxygen vacancies could be reduced by doping ions with smaller sizes, so as to improve the vacancy mobility [10]. Also, introducing the larger-sized

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ions (such as Gd^{3+} and La^{3+}) can cause elastic strain in the crystal structure due to the reduction of system energy.

In addition, synergistic effect also exists in the doping of materials. Multi-element doping can enhance the migration of oxygen vacancies, improve grain boundary conductivity, optimize ionic conductivity, and reduce electronic conductivity. The co-doping with Gd and Y elements resulted in $Ce_{0.8}Gd_{0.05}Y_{0.15}O_1$, whose conductivity at 500 °C was nearly three times higher than that of the Gd single-doped electrolyte, reaching 0.013 S/cm [11]. Zhu et al. prepared $Ce_{0.08}Gd_{0.16}Ba_{0.04}O_2$–δ-based electrolytes through the sol-gel method by adding BaO_2 after Ce doping in CeO_2, which reached a relative density of 98% and a conductivity of 2.46×10^{-2} S/cm at 700 °C [12]. Although the improvement effect of element doping on the macroscopic performance of batteries has been verified to be effective, the reaction mechanism of multi-ion doping still needs to be further investigated. In the future, the electrolyte performance can be a step further improved by precisely regulating the distribution structure of doped ions in the base material and reducing the migration barrier of oxygen ions.

3.2. Composite regulation of materials

Limitations can often be observed about the performance of a single material. However, by compounding two or more electrolyte materials with complementary properties to form solid solutions, heterojunctions, or composite ceramic structures, the ion migration capability and high-temperature stability of the materials can be effectively improved.

Liang et al. were the first to discover that after doping 35%–45% Al_2O_3 into LiI, the ionic conductivity of the material increased by two orders of magnitude in 1973 [13]. The mechanism may be related to the defect effect or interface effect which were formed between the doped phase and the base material. Choi et al. prepared a double-layer electrolyte consisting of 8μm YSL (yttria-stabilized zirconia) +40 μm GDC via dry pressing and spraying processes [14]. The peak power density of this composite structure could reach up to 218 mW/cm^2 at 600°C, which can be fully regarded as an example about the role of composite regulation in improving the performance of low-temperature SOFC electrolytes. However, the composite regulation technology of materials still faces many challenges: the ion conduction mechanism has not been fully revealed, the compatibility and long-term stability between materials need to be enhanced; in addition, the high cost of rare earth doping elements such as Gd and Sm leads to high preparation costs of SOFCs. These issues need to be focused on and addressed in future research.

4. Conclusion

Solid oxide fuel cells (SOFCs) have emerged as a promising option for clean power generation devices for their advantages such as high conversion efficiency, stability, and long service life. However, their demanding operation temperature leads to slow startup speeds and sintering issues, Impeding their applicability. The development of intermediate and low-temperature electrolytes has thus become a crucial pathway for the large-scale commercial application of SOFCs. Currently, mainstream electrolyte materials each possess distinct characteristics and limitations. For example, YSL-based materials, regarded as the most extensively studied electrolytes, exhibit high ionic conductivity only under a high-temperature environment for large ion migration resistance caused by their spatial structure. CeO_2-based materials, benefiting from a crystal structure that helps the formation of oxygen vacancies, offer advantages such as high conductivity and good structural tunability; however, in fuel gas environments, cerium ions show a tendency to reduction, which

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reduces the open-circuit voltage and hinders battery performance. Perovskite-type electrolytes, with their strong structural solubility and high modifiability, have become a research hotspot but issues such as easy reaction with battery materials still need to be solved.

To address the above problems, future improvements can be focused on the following aspects: (1) Enhancing vacancy concentration and ionic conductivity while reducing electronic conductivity through ion doping. (2) Conducting in-depth research on the mechanism of multi-element doping effects. (3) More efforts on improving the sintering performance of materials and enhancing their compatibility with battery devices through interface regulation. (4) Investigating nanoscale materials and utilizing nanomaterial preparation processes (such as vacuum deposition, sol-gel method, and in-situ nanoparticle precipitation technology) to leverage the size reduction effect of nanomaterials for performance enhancement. (5) Optimizing thin-film manufacturing technologies to reduce film thickness, minimize ohmic resistance, and improve the mechanical properties of materials. Resolving these issues will greatly expand the potential and prospects for application and marketing of SOFCs.

Authors contribution

All the authors contributed equally and their names were listed in alphabetical order.

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